REPORT DOCUMENTATION PAGE

Form Approved OMB NO. 0704-0188

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1. REPORT DATE (DD-MM-YYYY)	2. REPORT TYPE	3. DATES COVERED (From - To)
03-06-2009	Final Report	1-Jul-2005 - 31-Oct-2008

4. TITLE AND SUBTITLE	5a. CONTRACT NUMBER
Final Report for Agreement W911NF0510405, "Trapping and cooling of polar molecules", 07/01/2005-10/31/2008	W911NF-05-1-0405
	5b. GRANT NUMBER
	5c. PROGRAM ELEMENT NUMBER
	611102

6. AUTHORS	5d. PROJECT NUMBER
D. DeMille	
	5e. TASK NUMBER
	5f. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAMES AND ADDRESSES	8. PERFORMING ORGANIZATION REPORT NUMBER
Yale University Office of Sponsored Programs Yale University New Haven, CT 06520 -	

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)	10. SPONSOR/MONITOR'S ACRONYM(S)
U.S. Army Research Office P.O. Box 12211 Research Triangle Park, NC 27709-2211	ARO
	11. SPONSOR/MONITOR'S REPORT NUMBER(S)
	48781-PH.1

12. DISTRIBUTION AVAILIBILITY STATEMENT

Approved for public release; federal purpose rights

13. SUPPLEMENTARY NOTES

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14. ABSTRACT

This project centered on the development of new methods to trap, cool, and detect polar molecules, with potential application across a broad range of subfields. Progress was made on several inter-related fronts. A new, simple and general source for slow and cold molecular beams was developed. This source was demonstrated to work, with several molecular species, in two regimes: one with low velocity but also low flux, and another with higher velocity but also orders of magnitude greater brightness. The latter source is particularly suitable for precision spectroscopy

15. SUBJECT TERMS

cooling trapping polar molecule

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	15. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE			David DeMille
UU	UU	UU	UU		19b. TELEPHONE NUMBER
					203-432-3833

Standard Form 298 (Rev 8/98)
Prescribed by ANSI Std. Z39.18

Report Title

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List of papers submitted or published that acknowledge ARO support during this reporting period. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

Number of Papers published in peer-reviewed journals: 0.00

(b) Papers published in non-peer-reviewed journals or in conference proceedings (N/A for none)

Number of Papers published in non peer-reviewed journals: 0.00

(c) Presentations

APS DAMOP Meeting, May 2008
ICTP Workshop on Quantum Phenomena and Information, May 2008
APS March Meeting, March 2008
European Science Foundation Research Conference on Quantum Optics, Feb. 2008

Number of Presentations: 4.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):   0

Peer-Reviewed Conference Proceeding publications (other than abstracts):

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):   0

(d) Manuscripts

D. I. Schuster, Lev S. Bishop, I. L. Chuang, D. DeMille, and R. J. Schoelkopf, Cavity QED in a molecular ion trap, arXiv:0903.3552v1 [cond-mat.mes-hall], Submitted to Phys. Rev. Lett. (2009).

Lincoln D. Carr, David DeMille, Roman V. Krems, and Jun Ye, Cold and Ultracold Molecules: Science, Technology, and Applications, arXiv:0904.3175, Submitted to New J. Phys. (2009).

Number of Manuscripts: 2.00

Patents Submitted

Patents Awarded

Graduate Students

NAME	PERCENT_SUPPORTED	
John Barry	0.25	
David Glenn	1.00	
FTE Equivalent:	**1.25**	
Total Number:	**2**	

Names of Post Doctorates

NAME	PERCENT_SUPPORTED	
Edward Shuman	0.25	
Daniel Farkas	0.13	
FTE Equivalent:	**0.38**	
Total Number:	**2**	

Names of Faculty Supported

NAME	PERCENT_SUPPORTED	National Academy Member
David DeMille	0.01	No
FTE Equivalent:	**0.01**	
Total Number:	**1**	

Names of Under Graduate students supported

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Student Metrics
This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: 0.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:...... 0.00

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Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):...... 0.00

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The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields:...... 0.00

Names of Personnel receiving masters degrees

<u>NAME</u>

Total Number:

Names of personnel receiving PHDs

<u>NAME</u>
Jessie Petricka

Total Number: **1**

Names of other research staff

<u>NAME</u>   <u>PERCENT_SUPPORTED</u>

FTE Equivalent:
Total Number:

Sub Contractors (DD882)

Inventions (DD882)

Statement of the Problem Studied

Over the past several years, there has been an explosion of ideas for scientific applications based on the properties of ultracold polar molecules.[1] These ideas build on the revolutionary advances in atomic physics arising from the ability to cool and trap atoms. Extending these techniques to polar molecules gives access to new degrees of freedom associated with the internal (e.g. rotational and vibrational) states of molecules. These degrees of freedom can be used as powerful tools for new forms of control over quantum systems. Of particular interest is the large electric polarizability associated with the low-energy rotational states of polar molecules. This can enable unprecedented control over the interactions between quantum particles, via the strong, long-range, anisotropic, and tunable electric dipole-dipole interaction. The possible applications of ultracold polar molecules that have been envisioned include quantum information processing;[2,3,4,5] new frontiers in physical chemistry;[6,7] novel many-body quantum systems;[8,9,10,11] new or improved types of sensors;[12,13,14] and a variety of precision measurements and tests of fundamental physics.[15,16,17,18]

At present, the most advanced methods for producing ultracold polar molecules rely on assembly of the molecules from laser-cooled atoms.[19,20] In parallel, many groups are investigating methods for *direct* cooling and trapping of polar molecules, which may open a route to larger samples and/or a broader range of species.[21,22,23,24] Our work in this grant focused primarily on implementing various elements of our recently-proposed scheme[25] for direct cooling and trapping of polar molecules. The ultimate goal is to bring a large sample of polar molecules to the ultracold regime. Briefly, our scheme consists of the following steps:

a) precool molecules by collisions with a cryogenic buffer gas, and extract the cold molecules in the form of a guided beam
b) slow and cool the molecules from the beam sufficiently to load them into a trap
c) sympathetically or evaporatively cool the molecules to ultracold temperatures

Here we discuss the progress made towards these goals during the grant period. In parallel with this effort, we also devised ideas for a new architecture for quantum information processing: the "molecule chip". These ideas are also described.

Summary of the most important results

Buffer gas-cooled molecular beams

We developed a new, simple, very general technique for producing intense beams of cold molecules with low velocity. The basic method (shown in Fig. 1a) and initial results (with work done in collaboration with J. Doyle's group at Harvard) were reported in Ref. [26]. Molecules are vaporized at high temperature by laser ablation of a solid target. The target is located in a cell filled with He buffer gas. The gas and cell walls are in thermal contact with a cryostat (held at temperature $T \sim 4K$ for our system). After a sufficient number of collisions with the buffer gas atoms, molecules come to thermal equilibrium at T. Finally, molecules escape through an exit aperture, forming a beam. Initial work at Harvard used a variety of atoms and molecules. At Yale we initially focused on the highly polar species SrO but have subsequently also produced beams of several other species, including ThO and free radicals such as SrF and BaF.

As discussed in Ref. [26], we confirmed the expected trade-off between high beam flux and low beam velocity. A larger exit aperture allows a greater fraction of molecules to escape into the beam. However, collisions with exiting He (which has a much higher thermal velocity than the molecules, because its mass is much smaller) in the vicinity of the aperture can boost the
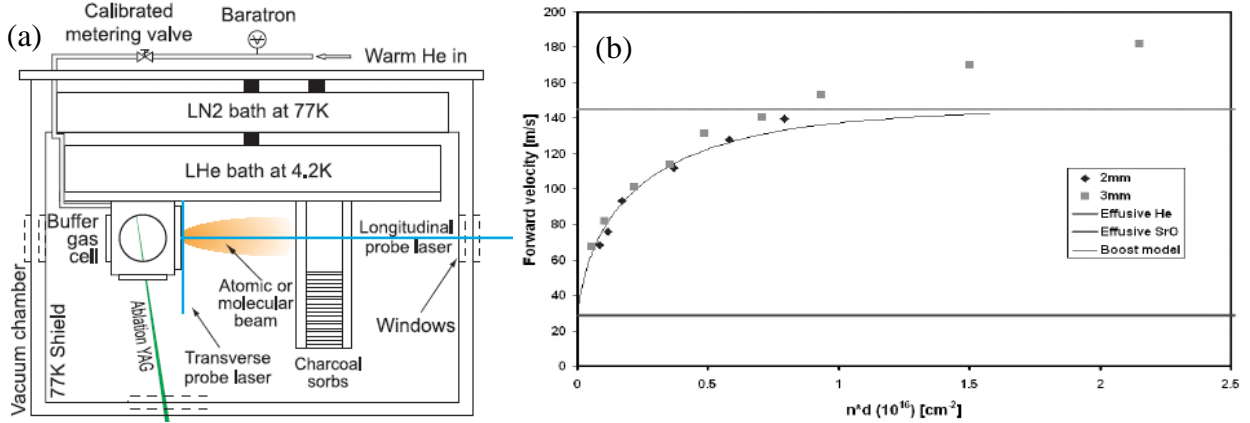
Fig. 1: Buffer gas-cooled beams. (a) Experimental schematic (b) Beam forward velocity as a function of (buffer gas density × aperture dimension), showing the "boost" effect. Points are data taken with two different aperture sizes; the curve is the result of a simple, 1-D kinetic model for the boost. Horizontal lines show thermal effusive beam velocities for He and SrO.

velocity of the molecules. Roughly, significant boost (e.g. doubling of the beam velocity) occurs when the exit aperture dimension d is made as large as the mean free path $\lambda = 1/n\sigma$, where n is the He density and σ is the collision cross-section (see Fig. 1b). Nominally requiring the molecular beam velocity v to satisfy $v < \sqrt{2}v_T$ [where $v_T \approx 30$ m/s is the thermal effusive beam velocity at $T = 4$K] sets an upper limit on the extraction efficiency ε for molecules into the beam, of $\varepsilon \approx 5 \times 10^{-4}$ (for an optimized, slit-shaped aperture).

After our collaborative work ended, Doyle's group began exploring variations on this beam-forming technology. They reported[27] a surprising result, namely: for large aperture sizes (i.e., in the regime of large forward velocity boost), ε could be dramatically higher than expected from geometric considerations alone (i.e., $\varepsilon \sim A/D^2$, where A is the aperture area and D is the characteristic length scale over which molecules thermalize in the cell). We have now qualitatively confirmed this behavior. Under typical conditions we reach $\varepsilon \approx 10\%$ for beams boosted up to $v_{He} \approx 150$ m/s. (Doyle's group has reported up to $\varepsilon \approx 50\%$.) We believe that this enhancement is due to hydrodynamic effects *inside the cell* (rather than in the beam as relevant for usual hypersonic beam sources), which sweep out the thermalized molecules in a "wind" of He atoms. Both groups have also observed that under typical conditions, the enhanced molecular beam remains cold ($T \sim 4$ K) in the transverse direction, so that the beam is substantially collimated (solid angle $\Omega \sim (v_T/v_{He})^2 \approx 0.03$ sr) directly out of the source. For example, with this technique we have produced beams of SrF with brightness B $\sim 3 \times 10^{11}$ mol/sr/state/pulse; pulse duration $t_p \sim 2$ ms; transverse velocity $v_\perp \sim 30$ m/s; forward velocity $v_\parallel \sim 150$ m/s; and rotational and translational temperature T ~ 4K.

These hydrodynamically-enhanced beams have velocity too high to be directly loaded into a trap. However, they appear to be useful in a variety of other ways, all related to the fact that their brightness (phase space density) is $\sim 1000\times$ ($\sim 100\times$) larger than has been demonstrated for other sources of condensable molecules. P.I. DeMille is already incorporating this type of source into two of his other experiments (tests of fundamental symmetries), where they will dramatically improve signal sizes. As described below, we also began using this beam as the starting point for new experiments on laser cooling of SrF.
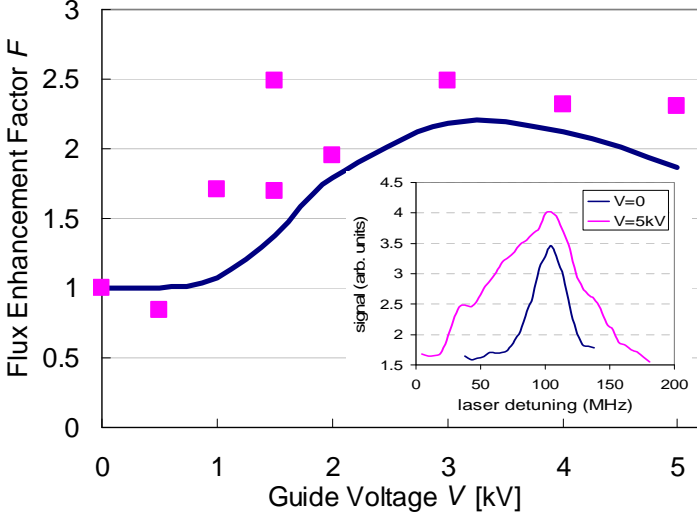
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Fig. 2: Demonstration of transverse beam guiding using an electrostatic quadrupole guide. These data reflect the population in the $J=2$ rotational state, for a 10 cm long guide. The main plot shows the ratio of signal with guide voltages on vs. off, vs. guide voltage. Points are data; the curve is from numerical simulations. The most strongly guided sublevel ($J=2$, $m=0$) is enhanced by a factor of >8 at the optimal field. The inset shows a scan of laser frequency with voltages on and off. The line broadening results from guiding of molecules with large transverse velocities.

In order to efficiently transfer molecules from the vicinity of the buffer gas cell (at $T \sim 4K$) to a UHV trapping region (at room temperature), it may be advantageous to transversely guide the beam using an electrostatic quadrupole field. We have seen the expected enhancement in downstream flux for a 10 cm long guide (see Fig. 2).

Finally, we note that the various diagnostics we perform on these molecular beams has required an unusual degree of flexibility in and control over laser operating parameters. For example, we found it necessary to develop an in-house capability for constructing tunable external-cavity diode lasers over a wide range of wavelengths (405-1020 nm). We can now produce such lasers from scratch within a few days. We also developed the capability to probe very small molecular absorption signals ($<10^{-4}$ absorption in a single ~ 2 ms duration beam pulse, with S/N$>$10) using FM spectroscopy. Finally, we developed a novel technique for frequency tuning and locking to arbitrary molecular lines. Here, the length of an optical cavity is locked by reference to a stabilized HeNe laser. The diode laser is then offset-locked to the cavity. The offset frequency is generated with an acousto-optic modulator (AOM), which can tune the offset continuously by more than one free spectral range (FSR) of a short "re-entrant mode" cavity[28] (FSR $\sim$ 90 MHz). To tune further, we instantaneously "jump" the offset backwards by one FSR, where the lock automatically recaptures; then again advance the offset. This "optical frequency ratchet" allows rapid, long-range tuning ($>$50 GHz total, at rates $>$1 GHz/s) with accuracy at the 1 MHz level (limited by reference HeNe stability). The technique can be extended to lock and tune multiple lasers anywhere in the range 400-1100 nm, with a single stabilized cavity. A paper describing this method is being prepared for submission to Rev. Sci. Instrum.

Development towards a microwave trap for molecules

The basic ideas underlying our proposed microwave trap for polar molecules were outlined in Ref. [25]. The microwave trap has a several attractive features: large trap depth and volume for efficient loading; the capability to trap absolute ground-state molecules, and hence suppress inelastic collisions; and enhanced elastic collision rates (and hence evaporative and sympathetic cooling efficiency) through electric polarization of the trapped molecules. To realize this vision we have undertaken to construct a microwave resonant cavity with high Q ($\sim 10^5$), an open geometry for optical access, and circular polarization (to avoid excitation of molecules to higher rotational states[25]), with high drive power ($P_{in} \sim 2$ kW). We use a Fabry-Perot microwave

Quality Factor (10^4)

10
8
6
4
2

◆ 30 cm sq mirror
■ 15 cm sq. mirror

Semi-Confocal

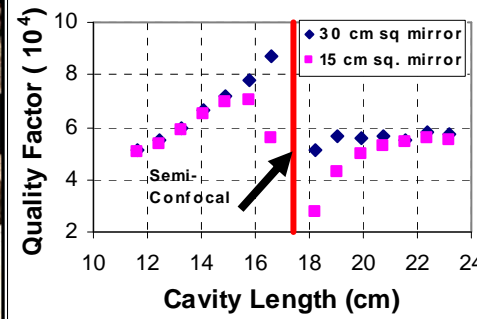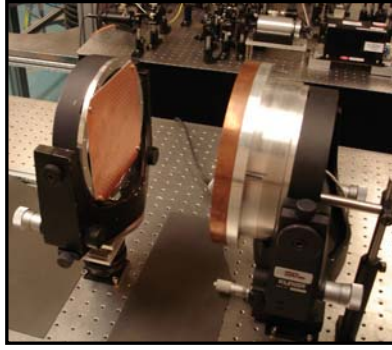10 12 14 16 18 20 22 24

Cavity Length (cm)

Fig. 3: Microwave cavity development. Left: a low-power test cavity with PCB input mirror. Right: cavity Q vs. mirror spacing, for two different input mirror sizes. Q plummets near the confocal geometry because of transverse mode mixing.

resonator, with input coupling through an array of sub-wavelength sized holes in a thin copper reflector.[29] Here we outline our progress in this direction.

We have extensively tested low-power prototype cavities, in order to optimize the properties of the final high-power system. For these tests, we primarily used input couplers fabricated from circuit boards with machined hole patterns. Mode matching was achieved by launching a Gaussian beam from a scalar horn, then focusing with either lenses or mirrors. Polarization control was achieved with waveplates fabricated from plastic plates with machined sub-wavelength groove patterns, and polarizers constructed from ordinary electrical ribbon cable. All these devices had to be custom fabricated, since quasi-optical elements at our wavelength (1.6 cm) are not standard. The beam mode was measured using a "beam profiler" constructed from a small pickup horn on a computerized two-axis stage with ~18" travel in each direction.

With these tools, have understood many limitations of these quasi-optical techniques and also optimized the design for the ultimate cavity and beam delivery system. For example, we ultimately achieved near the theoretical Q (~5×10^4 observed vs. ~7×10^4 computed) for an optimally-loaded cavity (see Fig. 3). We also found that the desired polarization and mode purity would be very difficult to achieve with quasi-optical elements. Hence, we ordered a custom-built feed horn for optimized coupling to the cavity. This horn includes, on its input, a "squashed-plate" waveguide circular polarizer; its output beam diameter is designed so as to match the required mode at the input to our high-power cavity.

The initial version of this circular polarizer performed far worse than its specification. (This problem was ultimately traced back to a bug in a standard commercial EM modeling package, HFSS from Ansoft!) Although the current polarizer meets our original specification, this problem induced us to calculate the effect of imperfect circular polarization on the molecules in our trap. We find that, with the cross-polarization level seen now, the maximum useful trap depth will be limited to ~1K, versus ~3K as originally anticipated. (The required input power is hence "only" 1.2 kW). This does not appear to be a significant problem as we move forward.

Our final trap will use a thermally conductive ceramic (BeO) substrate for the input mirror, in order to dissipate the deposited heat. We have also performed extensive modeling and tests related to the thermal management issues associated with the input mirror. In particular, we have identified a robust means for attaching water cooling lines to the substrate, with good thermal conductivity and adequate mechanical strength. This consists of vapor-depositing Cu on the edges of the substrate (just as needed on the front surface for the mirror itself), and then using a low-temperature braze alloy to attach copper cooling blocks housing the water lines.

Calculation of dissipative trap loading efficiency

As described in our original paper,[25] have investigated the use of a dissipative process for loading the microwave trap. This scheme is based on optical pumping from a weak-field seeking
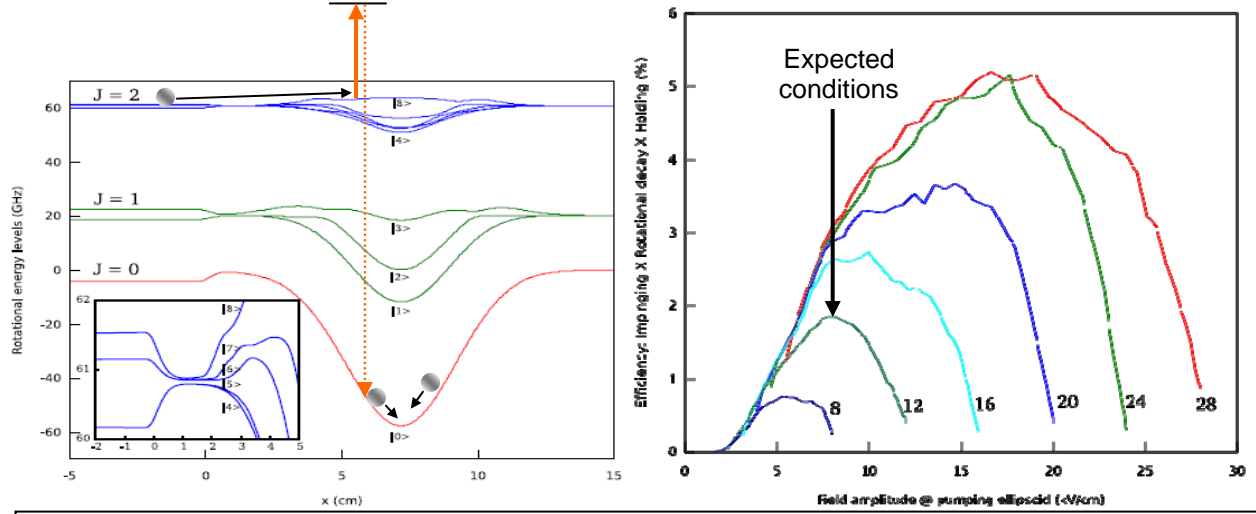
Fig. 4. Calculations of dissipative trap loading. (a) Molecular energy levels vs. position along a trajectory through the center of the trap. Molecules are guided into the region in the *J*=2, *m*=0 sublevel (labeled |8> here), then optically pumped (orange) near the center, into the *J*=0 trapped state (|0>). The inset shows adiabatic evolution of the initial state as it enters the trapping fields. (b) Efficiency for capture of molecules as a function of the microwave field amplitude where optical pumping takes place. Different curves correspond to different maximum field values, shown in the labels; we anticipate E_{max} ~ 12 kV/cm (arrow). The efficiency shown here should be multiplied by a Franck-Condon factor (~0.3 max for SrO) to get the final value for the trap loading efficiency.

excited rotational state, into the trapped strong-field seeking absolute ground state. This protocol (shown in Fig. 4a) has several advantageous features compared to the "trap-door" loading schemes typical of current molecular traps. It acts as a one-shot cooling process, in which the original longitudinal extent of the beam (typically ~10 cm for our conditions) is compressed into the trap volume (<1 cm here), hence increasing the phase-space density. It slightly slows the beam just before loading, hence increasing the captured fraction. Finally, it allows for multiple loadings of the trap. We have now performed detailed calculations to determine the maximum loading efficiency. Briefly, the key control parameter is the tuning of the optical pumping laser, which (because of the spatially-dependent Stark shifts in the trap) defines a thin ellipsoidal shell in space where molecules are pumped into the trapped state. Making this shell larger increases the spatial fraction of beam flux addressed, but increases the energy at which molecules "appear" in the potential felt by the trapped level. Hence, for a given maximum trap depth and input beam velocity distribution, there is some laser frequency which optimizes the number of trapped molecules. For typical expected conditions (see Fig 4b), we predict ~0.5% efficiency of capture from our guided beam. We note that for this calculation, it was crucial to consider the (adiabatic) evolution of the molecular states as they are transferred from the guiding (static field) to trapping (microwave field) regions. A paper describing this protocol and associated calculations is being prepared for submission to Phys. Rev. A. We note also that a similar dissipative loading scheme using atoms was recently demonstrated.[30]

A new architecture for quantum information processing: the "molecule chip"

During this grant period, we devised a new architecture for quantum information processing. Our work built on the recent spectacular success of R. Schoelkopf's group in developing "circuit QED" based on superconducting qubits in microwave stripline resonators (MSRs).[31] The new idea consists of trapping polar molecules just above the surface of a substrate containing an MSR
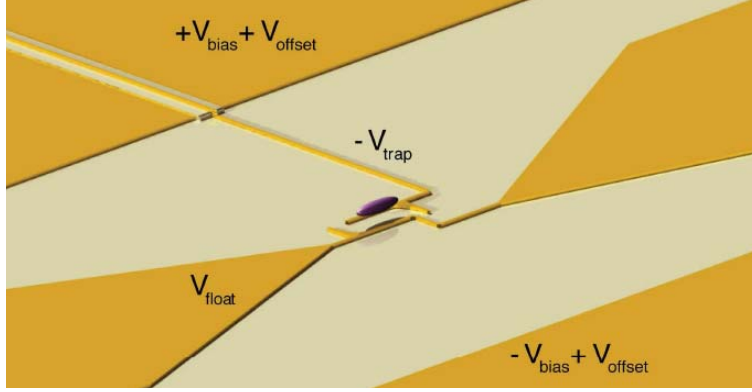
Fig. 5: Depiction of the proposed system for integrating molecular qubits with the circuit QED. The large structures are superconducting traces comprising a stripline resonator (only a small part of the total length is shown). The smaller features at the center create electrostatic potentials suitable for trapping polar molecules in the purple region.

(Fig. 5). Appropriate traps can be created by applying suitable voltages to electrodes patterned on the substrate. With reasonable parameters, the traps can be made quite deep (~1 K), making it conceivable to load from a simple source such as our cold molecular beams. Coupling of the molecule's motion to the MSR can provide a mechanism for dissipation and hence cooling of the trapped molecule to extremely low temperatures (~μK), which in turn provides strong localization of the molecules.

The system of ultracold trapped molecules, coupled to microwave photons in a MSR, combines many of the best features of atomic and condensed matter methods for quantum information processing. This system could provide fast, purely electrical means for gate control and high-fidelity qubit readout, plus transparent scaling properties based on micro-fabrication techniques. At the same time, it combines the inherent reproducibility of single-particle qubits and in addition (according to projections) the extraordinary immunity to decoherence that is characteristic of atomic systems. We published two papers on this system. Ref. [4] describes the ultimate vision of this scheme: individual trapped molecules cooled to the ground state of their motion, with simple gate operations and very low decoherence rates. Ref. [5] describes a simpler scheme with somewhat less favorable properties. It involves trapping ensembles of molecules in larger traps, and coupling these molecules to a Cooper-pair box qubit via microwave photons in a common MSR. The molecular ensemble acts as a (relatively) long-lived quantum memory coupled to a fast processor based on superconducting qubits.

Scheme for laser cooling and slowing of SrF molecules

Two recent papers[32,33] have outlined potentially viable techniques for laser cooling and trapping of molecules. We devised and began to implement a method which combines the best features of these proposals along with convenient
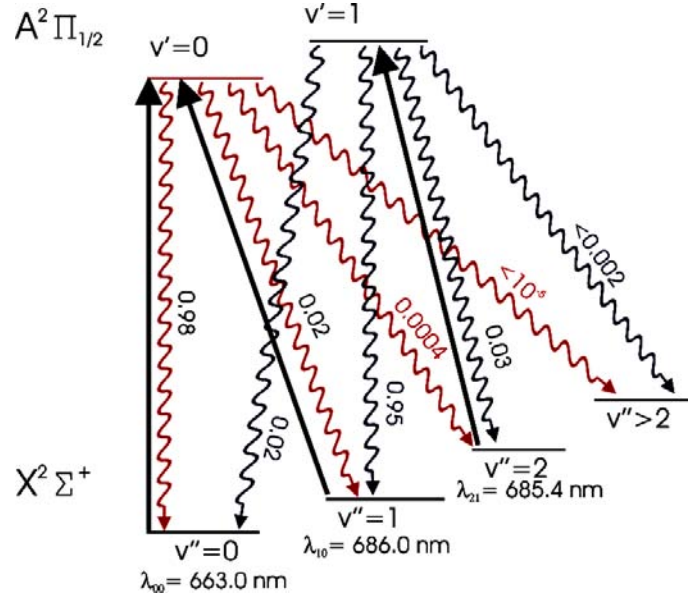

Fig. 6: Electronic and vibrational level structure of SrF relevant to laser cooling. Black upward arrows indicate laser-driven transitions, with wavelengths $\lambda_{v'',v'}$ as indicated. Wavy downward arrows indicate spontaneous emission paths, with Franck-Condon factors as indicated.

technological parameters, leading to what we believe is a fully realistic—indeed, even remarkably simple—scheme for laser cooling and slowing of SrF molecules. The technique uses our existing buffer-gas beam technology to provide molecular beams of high flux that are already moderately slow and cold. After application of laser cooling and slowing to this beam, the resulting sample of molecules should provide an ideal starting point for subsequent trapping and further cooling.

The primary obstacle to laser cooling of molecules has been the absence of closed cycling transitions, due to decays to unwanted rotational and vibrational sublevels. As in Refs. [32,33], our scheme relies on the choice of a molecule with highly diagonal Franck-Condon factors (FCFs) in order to suppress decays to excited vibrational states. Fig. 6 shows the relevant vibrational transitions in our initial target species, SrF. The $X(v=0) \leftrightarrow A(v'=0)$ transition is substantially closed, with ~98% branching ratio from the excited state. The off-diagonal FCFs for decays of the type $A(v'=0) \rightarrow X(v''\neq 0)$ diminish very rapidly as v'' increases.[34] By repumping only from the first two excited vibrational levels, $X(v''=1,2)$, we calculate that it will be possible to scatter enough photons to bring SrF molecules in our hydrodynamically-enhanced buffer gas beam to a complete stop.

The rotational branching can be kept under control by cooling on a transition of the type $N=1 \rightarrow N'=0$.[33] Here selection rules ensure that the excited state can only decay back to the original rotational state, so that only one laser is required per vibrational level. However, for any fixed laser polarization the molecule will be pumped to dark Zeeman sublevels (i.e., not excited by the laser). These can be "remixed" into the laser cycling transition by appropriate application of state magnetic and/or oscillating electric fields. For SrF in our buffer-gas cooled beam, we find that only 3 lasers (plus a handful of electronic modulation frequencies, see below) will be sufficient for laser cooling to ultracold temperatures and slowing to rest. The necessary wavelengths for the cooling ($\lambda_{00} = 663$ nm) and repumping ($\lambda_{01} = 686$ nm, $\lambda_{12} = 685$ nm) transitions are
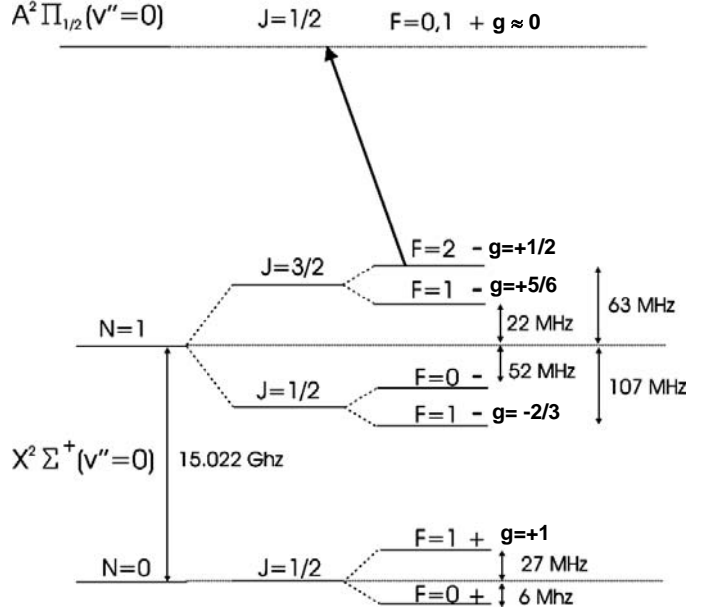

Fig. 7: Spin-rotation and hyperfine structure in SrF. Rotational angular momentum **N**, $\mathbf{J} = \mathbf{N} + \mathbf{J_e}$ (where $J_e = \frac{1}{2}$ is the electronic angular momentum), total angular momentum $\mathbf{F} = \mathbf{J} + \mathbf{I}$ (where $I = \frac{1}{2}$ is the nuclear spin of ^{19}F), parity $P = \pm 1$, and magnetic g-factor are shown for all relevant levels. The upper electronic state (A $^2\Pi_{1/2}$) has negligible hfs splitting. The nominal cooling transition is shown (arrow). Electric dipole selection rules allow transitions with $\Delta F = \pm 1, 0$ and change in parity ($P_iP_f=-1$). Hence only N=1 sublevels are accessed in decay of the upper state; however, all hfs sublevels are populated with similar probabilities. Our cooling schemes are designed to address all N=1 hfs sublevels, with similar efficiency. Microwaves near resonance with the $N=1 \leftrightarrow N=0$ rotational transition at 15.0 GHz can be used for dark state remixing (see text).

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available with sufficient power from standard diode lasers. Because the A-X transition of SrF has short lifetime (τ_A = 24 ns) and correspondingly broad natural linewidth ($\Gamma = 2\pi \times 7$ MHz), strong photon-scattering forces can be obtained for SrF with laser technology (power, linewidth) similar to that used for laser cooling of alkali atoms. Only moderate additional complexity is added to provide the necessary electronic modulations for hyperfine state repumping (see below).

All relevant energy levels of SrF have previously been measured to high accuracy (typically <300 MHz for electronic transitions, and <1 MHz for hyperfine/rotational splittings),[34] so that no spectroscopic studies are necessary to proceed with the laser cooling experiments. As in the case of CaH pursued by DiRosa,[32] in SrF the ground state has both an unpaired electron spin and an unpaired nuclear spin (from ^{19}F). The resulting spin-rotation and hyperfine substructure (hfs) must be taken into account, and it introduces some additional complexity to the laser cooling scheme (see Fig. 7).

Fig. 8: Laser and molecular hfs spectra for the pure laser current modulation scheme. The laser spectrum is shown in red; molecular lines (with power-broadened width $\gamma \approx 10$ MHz $\approx \Gamma\sqrt{2}$, corresponding to saturation parameter $\kappa = 1$) are in black. For clarity of presentation, each hyperfine line is shown with the same amplitude. For each separate molecular line, a laser sideband is detuned to the red of the transition by an amount varying from $(0\text{-}2)\times\Gamma$. In all cases, the scattering rate from blue-detuned lines is <1/50 the rate from red-detuned lines.

Effective laser cooling of SrF requires repump from all hfs sublevels of the N=1 rotational state. This can be accomplished with any of several standard techniques for radiofrequency modulation of lasers such as acousto-optic modulators, electro-optic modulators, or direct laser diode current modulation (see Fig. 8). Similarly, several schemes for remixing dark Zeeman sublevels into the cooling cycle appear viable. These include magnetic field mixing with a transverse B-field[35], polarization modulation of the cooling lasers[35], and or Stark remixing with a microwave electric field near resonance with the N=1↔N=0 rotational transition. In all cases the relevant technical parameters seem entirely feasible.

In our first steps towards laser cooling of molecules, we observed clear evidence for cycling fluorescence in SrF. Here only the X(v=0)→A(v=0) transition was driven, and the laser sideband frequencies are set near resonance. We began with an unmodulated laser, linearly polarized and tuned to the X(N=1,J=3/2, F=2)→A(J=1/2) line, and monitor decay fluorescence at the same wavelength. Next we monitored the change in the fluorescence signal upon addition of 40 MHz sidebands, and/or a transverse magnetic field. The results are shown in Fig. 9. Our results are consistent with ~10 photon scattering cycles, but a quantitative analysis (which must take into account power broadening, different velocity classes in the Doppler distribution, etc.) has not yet been performed. However, we can see no way to explain the dramatic increase in signal,
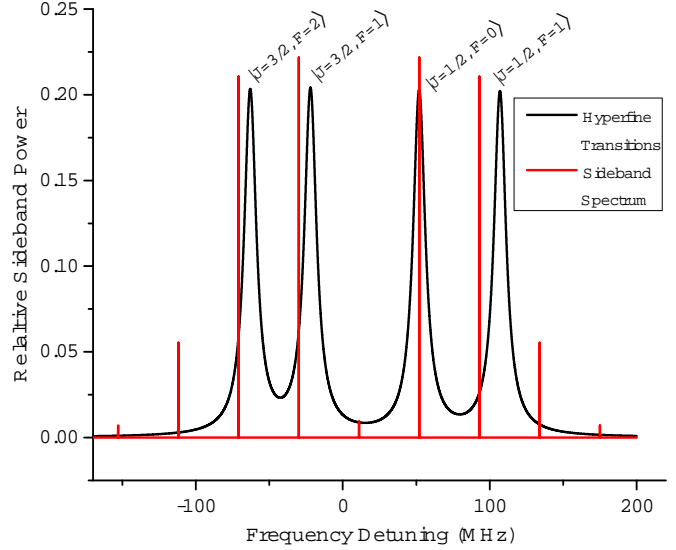
depending upon application of *both* hfs repumping sidebands and a transverse magnetic field, without a substantial number of photon scattering cycles.

Experimental development of molecule chips

Our initial theoretical investigation of the "molecule chip" architecture uncovered a central obstacle to moving forward experimentally. In order to trap molecules sufficiently close to the chip surface to couple significantly to the stripline resonator, it will be necessary to deliver molecules to the chip with phase space density orders of magnitude higher than is available in any current experiment. While this provides one of the motivations for the microwave trap work, it makes direct experimental progress towards a molecule chip difficult. As an alternative route, we began exploring the possibility for trapping *molecular ions* rather than neutral molecules in the first generations of molecule chips. The approach of using molecular ions rather than neutrals for the chip system has a number of attractive features. These include:

● Simple trap loading. For molecular ions, the trap-loading problem appears entirely tractable. With molecular ions, collisions
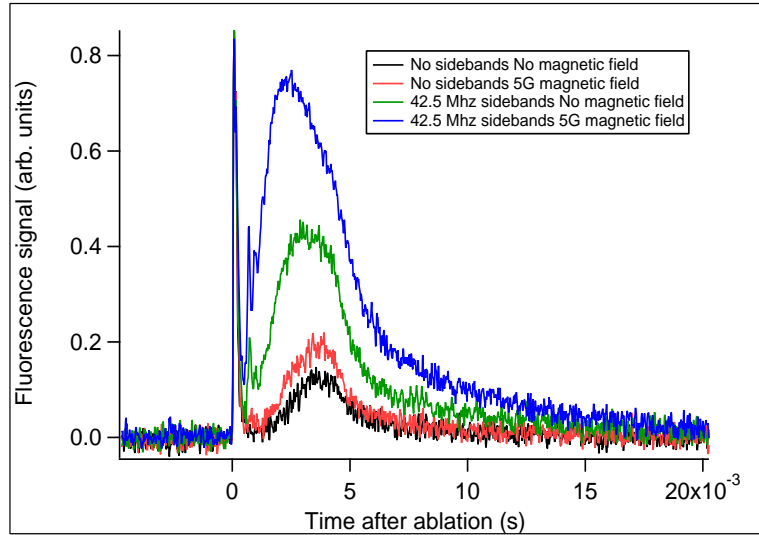

Fig. 9: First demonstration of cycling fluorescence in a diatomic molecule. The plot shows fluorescence signals from the A-X transition in SrF, as a function of time after the laser ablation pulse that initiates the SrF molecular beam. The black trace shows the signal for a single laser frequency, linearly polarized and tuned to the X(N=1, J=3/2, F=2)→A(J=1/2) line. The red trace shows a small increase in signal (~20%) on application of a transeverse magnetic field $B_\perp \approx 5$ G. The green trace shows a substantial increase of signal (~4×) on application of sidebands near resonance with all N=1 hfs sublevels but no B-field (similar to the situation in Fig. 8). Finally, the blue trace shows a further (~2×) increase in signal when the B-field is applied at the same time as the sidebands. The integrated signal size in the final configuration is ~10× larger than for the initial configuration. We believe the substantial increase in signal in the presence of $B_\perp$, but only when sidebands are present, can only be explained by cycling in the laser transition.

with buffer gas are a viable means for providing the dissipation needed for trap loading. (For neutrals this is impossible, since the small trap dimensions allow only static traps for weak-field seekers, i.e. rotationally excited states that would be rapidly quenched by collisions with the buffer gas.) In addition, the trap depths for ions are dramatically larger than for neutrals, and are essentially independent of the internal state of the molecule, unlike for neutrals. Finally, we note that atomic ions are now routinely loaded into sub-mm scale chip traps. Yale postdoc David Schuster spent several months in Chuang's lab at MIT, learning their methods for fabricating, loading, and detecting atomic ions on a cryogenic chip.[36] Based on this experience, we anticipate

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no difficulty fabricating a modest initial experiment with ~50,000 ions trapped in a 1 mm^2 × 1 cm linear trap, at T=1-4 K.

● Easy detection. According to our estimates, it should be possible to detect the anticipated sample of molecular ions simply by measuring absorption of resonant microwaves transmitted along a stripline located under the ion trap. This can be thought of as classic microwave spectroscopy (in the spirit of Townes and Schawlow[Error! Bookmark not defined.]), scaled down to the size of a chip. This method appears to scale naturally from use with large ensembles at modest temperatures as in our initial experiment, down to use with single molecules at low temperatures (<100 mK). In addition, the presence of ions of the desired species can be easily determined by releasing the trap and detection with time-of-flight mass spectrometry.

● Decoupling of internal states from trapping potential. The molecular ion's charge effectively shields the internal dipole moment (associated with its rotational motion) from the low-frequency electric fields associated with the trap potential. We crudely estimate that very long decoherence times (>1 sec) could be achieved for single molecules at the lowest temperatures. Even for the modest initial experiment, the estimated decoherence time could be in the ms range.

● Convenient species. Our first experiments will use the molecular ion $CaCl^+$. This has a fairly well-known rotational constant, with J=0→J=1 transition frequency 10 GHz.[37] The dipole moment is extremely large, μ = 11.7 Debye.[38] There is also a stable precursor for production via ablation, $CaCl_2$. Doyle's group has seen high-yield production of CaF neutrals by ablation of the chemical analogue CaF_2; hence we expect that CaCl will be produced in abundance, and if necessary can be photoionized by a subsequent REMPI process for ion trap loading.[39]

● Proof-of-concept demonstation. Chuang's group has recently demonstrated the ability to trap $CaCl^+$. In this proof of concept experiment, various ion species were co-loaded into a room-temperature ion trap, by simultaneous ablation of different target materials including $CaCl_2$. The ion cloud ensemble temperature was probed by standard laser-induced fluorescence from Sr^+ atomic ions. By modulating the trap electrode voltages at the motional frequency of trapped ions, it is possible to parametrically heat the ion cloud. Since the motional frequency depends on the ion's mass, this provides a means for mass-spectroscopic identification of all trapped species. Chuang's group saw clear evidence for trapping of $CaCl^+$ along with Sr^+ using this approach.

All in all, we believe that using trapped molecular ions will make it possible to explore many features of the coupled molecule/microwave stripline system, even while the development of ultracold neutral molecule sources progresses. The system appears potentially useful as a tool for rotational spectroscopy on molecular ions (for which there is typically very little data available), and may enable methods for detecting small numbers of molecular ions with purely electrical techniques and high species selectivity. We are have submitted a paper on the basic features of the molecular ion chips to Phys. Rev. Lett.

Summary and conclusions

We have made substantial progress in the development of intense, slow and cold molecular beams; towards methods for futher cooling and trapping of polar molecules in these beams; and in ideas for hybrid quantum information processing devices based on molecules trapped near the surface of superconducting chips. The future of all these methods appears promising.

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